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## Magnetic ordering in Fe/X/Gd (X=Cr, Pd) superlattices

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**Abstract.** We have studied the influence Cr and Pd spacers on structure and magnetic ordering in Fe/Gd superlattices. The insertion of Cr and Pd spacers in Fe/Gd has been found to cause structural changes in Gd layers. In particular, an additional fcc Gd phase appears in Fe/Cr/Gd along with the main hcp Gd phase, while there is amorphous Gd structure in Fe/Pd/Gd. By combining SQUID magnetometry and polarized neutron reflectometry we have shown that interlayer Fe-Gd exchange coupling through Pd spacers is much stronger than through Cr one.

### 1. Introduction

Metallic multilayers composed of alternating nanosized magnetic and nonmagnetic layers represent a new class of artificial materials that exhibit unique magnetic and magnetotransport properties. Of particular interest are multilayers consisting of layers of both rare earth (RE) and transition metals (TM). Heavy RE metals are characterized by large spontaneous magnetic moments, which are substantially temperature-dependent, and by complex magnetic ordering, which is due to the indirect RKKY interaction of 4f-electron magnetic moments [1]. Ferromagnetic TMs (Fe, Co, Ni) are characterized by lower spontaneous magnetic moments; however, their Curie temperatures are much higher than room temperature [2]. Superlattices composed of both RE and TM layers are frequently displaying a good deal of novel properties not observed in bulk RE and TMs [3-5].

Fe/Gd is an example of system, in which there appears an induced spontaneous magnetic moment in RE near Fe-Gd interfaces due to proximity effects [6, 7]. The induced moment was found to be temperature-independent and as large as to  $7 \mu_B/\text{atom}$  far above the Curie temperature of bulk Gd  $T_c=293\text{K}$ , which opens ways to create room temperature rare-earth magnets with large magnetization [8]. The main drawback of Fe/Gd system is Fe and Gd magnetic moments are antiparallel due to antiferromagnetic Fe-Gd exchange coupling, so the total magnetic moment remains low. Several years ago it was suggested to modify the Fe-Gd interlayer exchange interaction via insertion of antiferromagnetic spacer Cr layers between Fe and Gd [9]. Cr is a spin-density-wave antiferromagnet, where in thin enough films there is a simple antiferromagnetic ordering of neighbouring monolayers. Since the Cr magnetic moments are coupled antiferromagnetically with both Fe and Gd moments, the



Fe–Gd ferromagnetic ordering can be realized in layered Fe/Cr/Gd nanostructures with odd number of Cr monolayers.

Modification of Fe–Gd magnetic ordering via inserting Cr spacer was confirmed experimentally with x-ray magnetic circular dichroism (XMCD) in Fe/Cr/Gd three-layer system with a Cr layer of five monolayers in thickness [9]. By measuring Gd and Fe XMCD signal it was found that the projection of magnetic moments on magnetic field is of the same sign for both metals. It does not mean that Fe and Gd moment are ordered ferromagnetically but the canted angle between them is less than 90 degrees. In our previous article, we have shown that interlayer exchange interaction in Fe/Cr/Gd oscillates with Cr layer thickness with a period of Cr 1.8 nm [10].

Dependence of RE-TM exchange coupling on Cr spacer thickness was studied for Co/Cr/Gd multilayers by G. Suciú and al. [11], who found that both bilinear and biquadratic exchange coupling are significant in the system. As follows from their results, there is a rapid decrease in bilinear coupling and strong increase in biquadratic one with Cr spacer thickness increase, so that for Cr spacer thickness more than 1 nm there appears a canted phase with an intermediate angle between Gd and Co magnetic moments. Ferromagnetic Gd-Co ordering was not found in Co/Cr/Gd for any Cr thickness.

Interlayer Fe-Gd exchange coupling through different 3d, 4d, and 5d transition metal spacers was studied theoretically by C. Autieri et al. [12, 13], who predicted ferromagnetic RE-TM coupling for several antiferromagnetic and paramagnetic spacers like Cr, Mn, W, Ta. RE-TM exchange coupling through paramagnetic spacers is presently much less studied experimentally. We mention the results of G. Suciú and al. [11] who detected strong bilinear coupling and relatively weak biquadratic one in Co/Pt/Gd multilayers for all Pt spacer thicknesses up to 3 nm. In our previous article [14] we reported on the magnetization behaviour of Fe/Pd/Gd superlattices, where inserting Pd (1.2nm) spacer between Gd and Fe layers allows one to suppress a spin-flop transition field down to 1.5 kOe.

In the present article, we report on magnetic state in Fe/X/Gd (X=Pd, Cr) superlattices determined directly with polarized neutron reflectometry for spacer thickness up to 1.2 nm. We show that inserting the spacer layer cause structural changes in Gd layers that affect their magnetic properties. Magnetic ordering is shown to differ essentially for different spacer types.

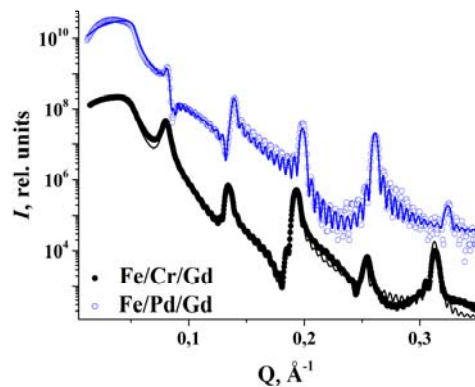
## 2. Experimental

Two series of [Fe(3.5 nm)/X(t)/Gd(5 nm)/X(t)]<sub>12</sub> X=Cr, Pd superlattices with t = 0.5-1.2 nm were grown with UHV magnetron sputtering at room temperature onto single-crystal (001)Si substrates. The growth was started with a 5 nm Cr buffer layer, followed by superlattice structure started with Gd layer. To protect the structure from oxidation, a 3 nm Cr capping layer was deposited at the top of the superlattice stack.

The structural characterization of the superlattices was performed with x-ray reflectometry and diffractometry using PANalytical Empyrean Series 2 diffractometer with Co and Cu K $\alpha$  radiation in parallel beam geometry. The parallel incident beam was formed using a parabolic W/Si mirror, beam height bounded by a slit being 0.08 mm. At the secondary beam we used Parallel Plate Collimator with Equatorial Acceptance 0.27 together with a plane graphite monochromator and a collimating slit with 0.1 mm equatorial aperture. The fitting experimental curves to model was done using the commercial PANalytical X'Pert Reflectivity software.

Figure 1 shows typical measured and model reflectivity curves, selected for two samples superlattices with [Fe/Cr(1nm)/Gd/Cr(1nm)]<sub>12</sub> and [Fe/Pd(1 nm)/Gd/Pd(1 nm)]<sub>12</sub> nominal composition. In the fit we considered a model of twelve identical [Fe/X/Gd/X] quadrolayers. Experimental curves were well described by a model structure with layer thicknesses of no more than 5% off the nominal values and rms roughness of order of one monolayer only.

Crystalline structure was found to be amorphous for all Fe/Pd/Gd/Pd samples but polycrystalline one for Fe/Cr/Gd/Cr. In the latter series Fe/Cr/Gd/Cr we detected as main phases hcp (0002)Gd and bcc (110)Fe ones. The insertion of a Cr layer between the Gd and Fe layers leads to structural changes in the Gd layers and appearance of the additional fcc phase in them along with the main hcp phase [15].

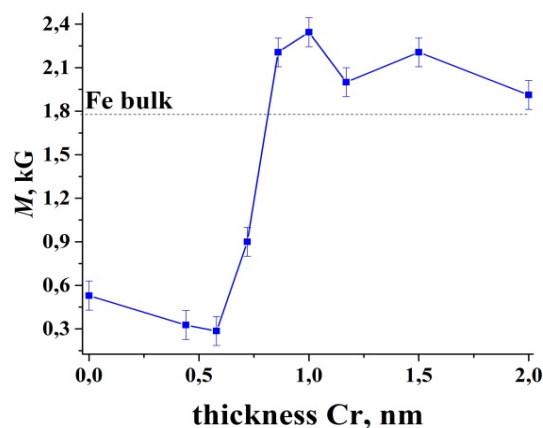


**Figure 1.** Reflectometry patterns measured for the  $[\text{Fe/Cr}(1 \text{ nm} / \text{Gd/Cr}(1 \text{ nm}))]_{12}$  and  $[\text{Fe/Pd}(1 \text{ nm/Gd/Pd}(1 \text{ nm}))]_{12}$  superlattices.

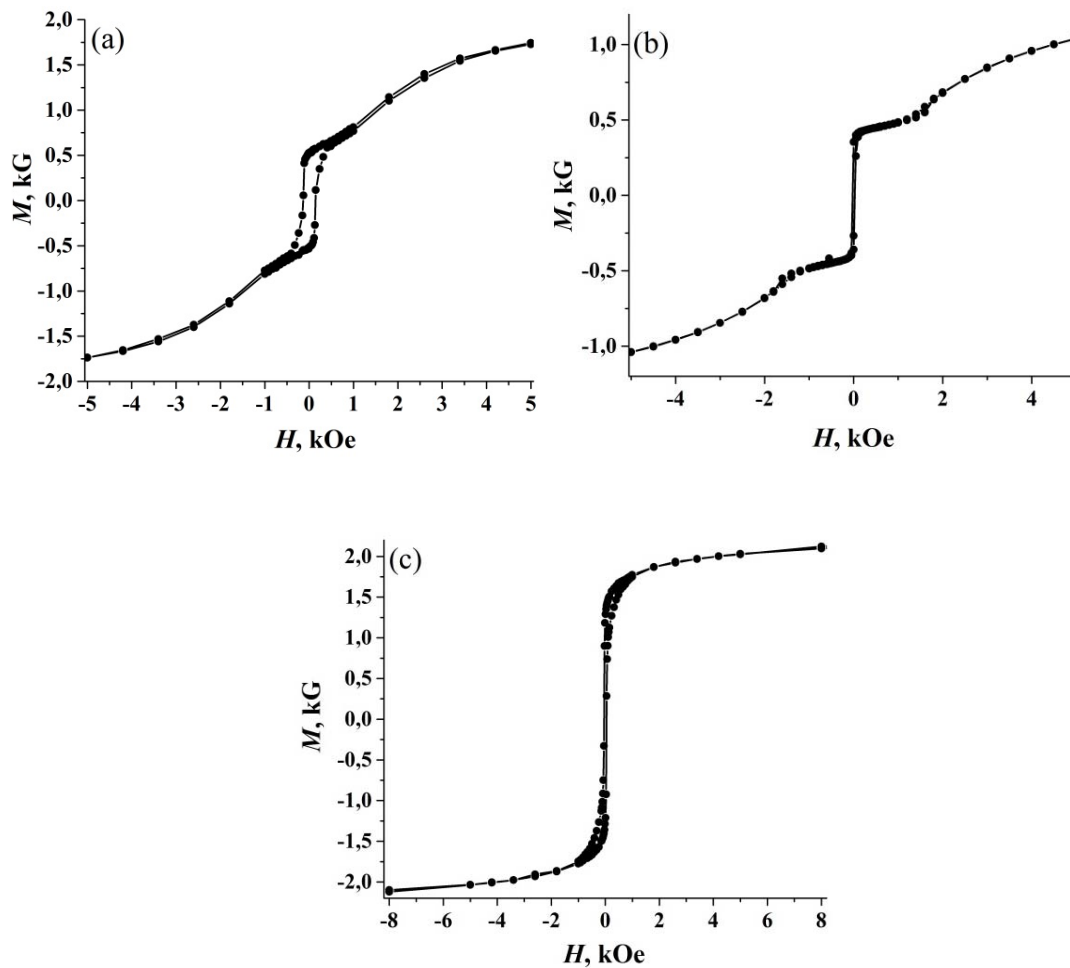
Magnetization measurements were performed at SQUID magnetometer with an in-plane magnetic field up to 60 kOe within temperature range from 15 and 300 K. Polarized neutron reflectometry experiments were performed at the NREX reflectometer situated at the FRM-II research reactor in the Heinz Maier–Leibnitz (Munich, Germany). The measurements with the full polarization analysis were carried out in magnetic fields of 0.5–4.5 kOe at temperature range 15–300 K using standard  $\theta$ - $2\theta$  geometry and with constant neutron wavelength of 4.3 Å.

### 3. Results and discussion

Figure 2 shows the dependence of the magnetization on the Cr spacer thickness in Fe/Cr/Gd superlattices obtained by SQUID magnetometer. The measurements were carried out at 15 K in a magnetic field  $H = 4 \text{ kOe}$ . The magnetization value was calculated by normalizing the magnetic signal to number of superlattice periods and Fe volume in one layer. We note that for Cr spacer thickness less than 1 nm the magnitude of the magnetization is essentially lower than that for bulk Fe, which implies that the projections of Fe and Gd magnetic moments on magnetic field are of opposite sign. As the Cr thickness increases further, superlattice magnetization becomes larger than that of bulk Fe and reaches 2.4 kG at  $t_{\text{Cr}} = 1 \text{ nm}$ , so Fe and Gd are ferromagnetically coupled or exchange coupling is absent. In the Cr thickness range investigated, magnetization oscillations are observed, which may be related to oscillations of the exchange interaction of Fe and Gd through the Cr layer.



**Figure 2.** The dependence of the magnetization of the Fe/Cr/Gd/Cr superlattices on the thickness of the Cr interlayer measured at  $H=4 \text{ kOe}$ ,  $T=15 \text{ K}$ .



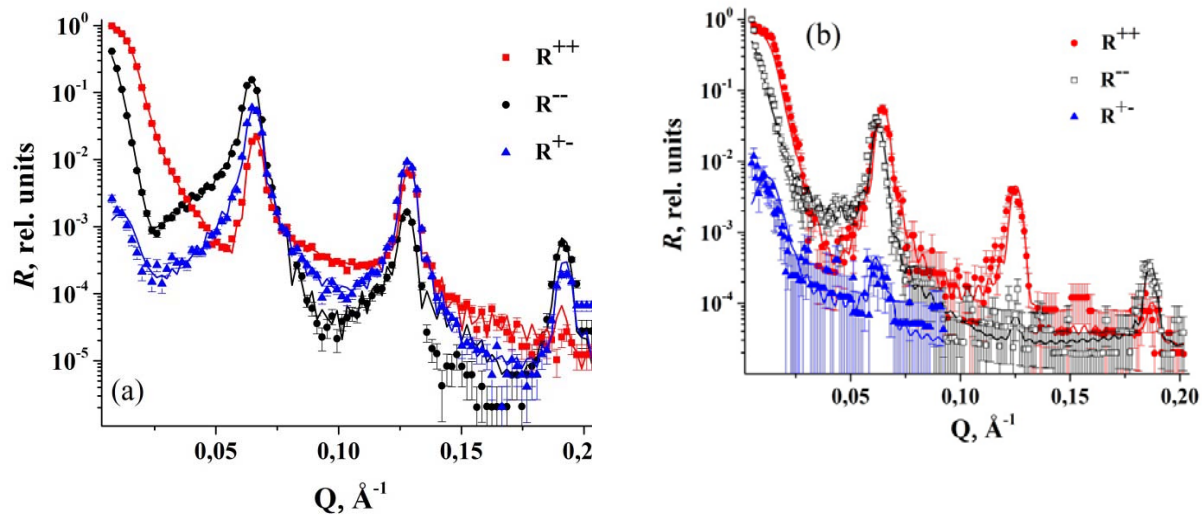
**Figure 3.** Experimental magnetization curve, measured for  $[\text{Gd}/\text{X}/\text{Fe}/\text{X}]_{12}$  multilayers with (a)  $t_{Cr}=0.6$  nm at  $T=100$  K, (b)  $t_{Pd}=1.2$  nm at  $T=15$  K and (c)  $t_{Cr}=1$  nm at  $T=15$  K.

In figure 3 are presented magnetization curves obtained for samples with thin Cr spacer layers up to  $\sim 0.6$  nm (a),  $\sim 1$  nm (c) and Pd spacer 1 nm. The curves in figures. 3(a) and 3(b) are typical of superlattices with strong bilinear and non-zero biquadratic interlayer exchange coupling [8]. The curve in figure 3c is typical for multilayers with almost ferromagnetic interlayer ordering and weak interlayer coupling. As follows from the pictures, for the first two samples in remanence there is non-collinear ordering between Gd and Fe. As magnetic field increases, in the sample with Cr spacer, a spin-flop transition occurs up to the field where saturation is not attained. In the sample with Pd spacer, one can observe first a “plateau” at the magnetization loop where Fe and Gd magnetic moments are aligned antiferromagnetically along and opposite to magnetic field. Next, as magnetic field increases a spin-flop transition starts to appear but the magnetization increases slowly as a function of the applied field before the transition begins; and in the high field the magnetization never seems to reach the saturation. In the sample with thicker Cr spacer (figure 3c), spin-flop transition does not occur, the magnetization change is likely to be due to domain walls movement inside Fe and Gd layers. The above different scenarios for magnetic behavior in Fe/Cr/Gd/Cr and Fe/Pd/Gd/Pd are due to different strength of Fe-Gd interlayer coupling through different spacers.

In order to study the magnetic ordering in Fe/Pd/Gd/Pd and Fe/Cr/Gd/Cr systems we used polarized neutron reflectometry experiment. It is best to focus on the low temperature measurement done at  $T=15$  K and  $H=4.5$  kOe. Figure 4 shows the reflectivity scans of samples (a)  $[\text{Fe}(3.5 \text{ nm})/\text{Pd}(1 \text{ nm})/\text{Gd}(5 \text{ nm})/\text{Pd}(1 \text{ nm})]_{12}$  and (b)  $[\text{Fe}(3.5 \text{ nm})/\text{Cr}(1 \text{ nm})/\text{Gd}(5 \text{ nm})/\text{Cr}(1 \text{ nm})]_{12}$ .

We fitted experimental curves to the model using GenX software [16]. In the fit we used thicknesses and rms roughness obtained from XRR and varied absolute values of magnetizations ( $M_{\text{Gd,Fe}}$ ) and tilting angles ( $\alpha_{\text{Gd,Fe}}$ ) between magnetization vector  $M_{\text{Gd,Fe}}$  and  $H$ . At  $H = 3.5$  kOe and  $T = 15$  K for sample Fe/Pd(1 nm)/Gd/Pd(1 nm) we observe strong increase of the intensity of spin-flip scattering which evidences increase of non-collinearity of the system. Best correspondence between model and experiment is obtained for the  $M_{\text{Gd}} = 1380 \pm 100$  G,  $M_{\text{Fe}} = 1200 \pm 100$  G,  $\theta_{\text{Gd}} = 347 \pm 5$  deg and  $\theta_{\text{Fe}} = 98 \pm 5$  deg. Such a highly non-collinear magnetic state is explained by competition of antiferromagnetic coupling and Zeeman energies (so called spin-flop transition). Since magnetic moment of Fe is smaller its magnetization vector will rotate at small fields antiparallel to the external field.

For the second system Fe/Cr/Gd/Cr the spin-flip scattering negligible quantity. Thus we have parallel alignment of  $M_{\text{Gd}}$  and  $M_{\text{Fe}}$  proving the absence of antiferromagnetic exchange coupling of Fe and Gd moments through the 1 nm Cr spacer. The magnetization at Gd interfaces is  $1130 \pm 100$  G, whereas the magnetic moment in the center of the layer is only  $1010 \pm 100$  G. The magnetic moment in Fe layers is independent of temperature and equals  $1621 \pm 100$  G. Note that the proximity effect is also observed in Fe/Gd superlattices and Fe/Cr(0.4 nm)/Gd/Cr at high temperatures [8, 17, 18].



**Figure 4.** Polarized neutron reflectivity curves for neutrons polarized along ( $R^{++}$ ) and opposite of the direction of the magnetic field ( $R^{--}$ ) for the Fe/X(1 nm)/Gd/X(1 nm) structure (a) X=Pd  $H = 3.5$  kOe, (b) X=Cr in the magnetic field  $H = 4.5$  kOe at a temperature of 15 K; symbols and solid lines correspond to experimental and calculated data, respectively.

#### 4. Conclusions

We have studied the influence of Cr and Pd spacers on structure and magnetic ordering in Fe/Gd superlattices. The insertion of Cr and Pd spacers in Fe/Gd has been found to cause structural changes in Gd layers. An additional fcc Gd phase appears in Fe/Cr/Gd along with the main hcp Gd phase, while there is amorphous Gd structure in Fe/Pd/Gd/Pd. By combining SQUID magnetometry and polarized neutron reflectometry we have shown that interlayer Fe-Gd exchange coupling through Pd spacers is much stronger than through Cr one. According to the polarized-neutron reflectometry data, the aforementioned structural changes are accompanied by a substantial (two-fold) decrease in the average magnetization of Gd over a wide temperature range.

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